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(54) RESIN COMPOSITION

(57)Abstract:

PURPOSE: To obtain a resin composition having excellent crack resistance in soldering and moisture resistance by blending mainly a modified reactional product obtained by reacting a specified polymaleimide with a silane coupling agent.

CONSTITUTION: This resin composition is manufactured by modifying (A) a polymaleimide containing ≥ 2 maleimide groups in one molecule (e.g. N,N'-ethylenebismaleimide) by reacting 100 pts.wt. of (A) with 0.1-25 pts.wt. of (B) a silane coupling agent of the formula (Y is univalent organic acid containing amino group; R₁ to R₃ are H, phenyl, OR (R is H or 1-5C alkyl), etc.; at least one of R₁ to R₃ is OR) (e.g. 3-aminopropyltrimethoxysilane) and blending the modified product with an epoxy resin containing ≥ 2 epoxy groups in one molecule and a hardener. Optionally the component A is mixed with a phenol resin, in advance.

R₁

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Y-S 1 - R₂

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R₃

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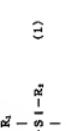
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CLAIMS

[Claim(s)]

[Claim 1] A polymaleimide compound and a general formula (1) which have two or more maleimide groups in one molecule (=ring 1)
[Chemical formula 1]



[An organic group of monovalence in which Y contains an amino group among a formula, R₁, R₂, and R₃. A hydrogen atom, a phenyl group, an allyl group of the carbon numbers 1-6. Or a thermosetting resin composition which contains as a subject a reaction-or-degeneration thing to which are OR group (R shows an alkyl group of a hydrogen atom or the carbon numbers 1-5), and at least one of R₁, R₂, and the R₃ comes to carry out the reaction of degeneration of the silane coupling agent expressed with, which is -OR group.

[Claim 2]The thermosetting resin composition according to claim 1 which made a polymaleimide compound contain phenol resin beforehand in [Claim 3]A resin composite by which an epoxy resin which has at least two epoxy groups being included in the thermosetting resin composition according to claim 1 or 2 and one molecule.

[Claim 4]A resin composite for semiconductor closure which contains the resin composite according to claim 1, 2, or 3 as a subject.

[Claim 5]A resin composite for semiconductor closure which contains a resin composite according to claim 1, 2, or 3 and an inorganic bulking agent as a subject.

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DETAILS AND DESCRIPTION

Detailed Description of the Invention

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[0001] Industrial Application [This invention is concerned with the resin composite excellent in heat resistance and dimensional stability and moisture resistance. In particular, it is related with a resin composite suitable for closing the semiconductor device or which solder heat resistance is required like a surface mount type semiconductor device.

[0002] Description of the Prior Art In recent years, the heat-resistant, crack-resistant, and damp-proof outstanding resin composite is called for in the field of an electric electronic component, a semiconductor, etc. For example, in the field of a semiconductor, it is the tendency for a packaging density to the

the laminating board which attaches parts with the miniaturization of an electronic component, apparatus, equipment and slimming down which lead this to become high, and the parts itself as especially in the field of multi-functionalization of multi-functionalization. The material which does this is strongly expected development of the resin composite excellent in heat resistance. As the high temperature solder in the soldering process to a semiconductor board. As the resin composite to such use, and what is called a resin composite for semiconductor exposure, conveniently. The epoxy resin represented by α -cresol novolac type epoxy resin and the resin which uses novolac resin and silica as the main ingredients that hardening agent are excellent in respect of a mobility and reliability, and serves as the mainstream in this field. ¹⁴Kiuchi

[00003] problem to be solved by the invention. However, speaking of a plastic molded type semiconductor device, it is difficult to change to a surface mount type semiconductor device by the flow of the above-mentioned high-density assembly-aligning. Unlike the conventional inserting type semiconductor device, in such a surface mounted type semiconductor device, the whole semiconductor device is put to the soldering temperature of the soldering furnace. By the way, the resin composite which holds the chip and the lead frame together is the epoxy resin, the melting point of which is relatively high. If the heat of the infrared heater at soldering temperature is intense, end it holds while the resin composite is still in the state in which the resin alone is relatively soft. Therefore, he who has an aversion of high-temperature soldering can not use the invention.

resin is to cool and harden, a crack occurs in sealing resin, and the reliability of a semiconductor device is guaranteed substantially.

magazine, the Nikkei Electronics June 13, 1988 item 114 - 118-page".

Therefore, a passage given in JP-H1-21335, JP-H2-254735A, and JP-D-32117A. This invention made glass transition temperature of sealing resin high, so the purposes of giving sufficient resin strength which overcomes the stress generated by expansion of moisture, and came in piles the research of the soldering joint which is applied to the main composite for closure. As a result, although intensity at the high temperature of an elevated temperature could be made high by application of mold resin and the crack resistance to the adhesive property with a semiconductor device, oxidation occurred in the surface of an element surface and resin by the thermal shock at the time of soldering, and there was a possibility that subsequent moisture resistance fail. The purpose of this invention is to provide the resin composite applicable to a surface mount type semiconductor device with which a reflow and flow soldering are made for closure by improving the soldering joint nature, the time of mounting, and the moisture resistance after mounting.

[0004] [Means for solving problem] In order to solve an aforementioned problem, as a result of repeating research

wholeheartedly, by denaturing a polymaleimide compound by the silane coupling agent which has an amino group, this invention persons find out that the adhesive property of the semiconductor device surface and resin improves, and hence to complete this invention. That is, this inventions are (1) polymaleimide compound and a general formula (1). [—Zing 2.]

The organic group of the monovalence in which Y contains an amino group among a formula, R_1 , R_2 and R_3 — a hydrogen atom, a phenyl group, and the alkyl group of the carbon numbers 1-6. If OR group (R shows the alkyl group of a hydrogen atom or the carbon numbers 1-6), at least one [and] of R_1 , R_2 and the R_3 — OR group — it is — the resin composite consisting of the thermosetting resin and the inorganic bulking agent, which make a subject the reaction of degeneration thing to which it comes to carry out the reaction of degeneration of the silane coupling agent expressed. (2) The thermosetting resin composition of the above-mentioned (1) description which made the polyimide compound contain phenol resin beforehand. (3) A resin composite by which the epoxy resin which has at least two epoxy groups being included in a thermosetting resin composition the above (1) or given in (2), and one molecule. (4) It is a resin composite for semiconductor device which contains the resin composite and inorganic bulking agent composed as a subject in the resin composite for semiconductor device which contains the resin polyimide compound used by this invention, it is a general formula (a), (2), (3) or (4). As a

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JP,06-157754,A [DETAILED DESCRIPTION]

hexoxy- α -phenyl and α -dimethylbenzyl phenoxyl- β -dihydro-1,3-butadiene, 1,3-butadiene-4-(4-maleimide phenoxyl)- α -phenyl and α -dimethylbenzyl phenoxyl- β -dihydro-1,3-butadiene, 1,3-butadiene-4-(4-maleimide phenoxyl- β -dihydro-1,3-butadiene) benzene, 1,3-butadiene-4-(4-maleimide phenoxyl- β -dihydro-1,3-butadiene) benzene, 1,3-butadiene-4-(4-maleimide phenoxyl- β -dihydro-1,3-butadiene) benzene, 1,3-butadiene-4-(3-maleimide phenoxyl- β -dihydro-1,3-butadiene) benzene, 1,3-butadiene-4-(3-maleimide phenoxyl- β -dihydro-1,3-butadiene) benzene, a general formula. (2) ³



the same time, the Fe^{2+} concentration was increased to 10 mM.

kind, may be mixed and they may be used. The silane coupling agent used for this invention is expressed in the above-mentioned general formula (1).

3-(2-methoxyethyl)propyltrimethoxylamine, N-(3-methoxyethyl)propylurea, N-(3-aminomethyl)ethyl trimethoxylamine, 2-(2-methoxyethyl) trimethoxylamine, a dimethylaminoethyl 3-piperazine dihydroxyethyl diethoxymethane, 3-piperazine propyltrimethoxylamine etc. are listed and these things are independent or are the result of degeneration of a polyimide compound and a saline coupling agent, a saline coupling agent carries out heating mixing of a polyimide compound and a saline coupling agent, and is performed. In this case, the common phenol resin and a saline coupling agent, and is performed.

The mixture ratio of a polyimide compound and a silane coupling agent, although it is the range of 0.1–25 mol-%, depends on the type of silane coupling agent used. The mixing ratio of silane coupling agents and restriction in particular does not have the mixing ratio of 100 mol-%, the same as that of a polyimide compound to 100 mol-%, the same as that of a polyimide compound to 100 mol-%.

loadings of a silane coupling agent are 0.1. A reaction of degeneration is insufficient for the case of less than a part, and it exceeds 25 copies, a silane coupling agent independent reaction will start and it will have an adverse effect on moisture resistance.

independent of the time which elapses after the resin has been applied. Compatibilizing agents such as phenol-formaldehyde, or epoxy resins, are especially useful in this respect.

The novolak type epoxy resin derived from novolak, hydroquinonebenzaldehyde, benzaldehyde, and methanol. Acetaldehyde, benzaldehyde, cresol, resorcinol, and methanol. Acetaldehyde, benzaldehyde, such as Guru oxal and alkoxanediol, and the aralkyl type epoxy resin which is a resultant of aldehyde, such as Guro above-mentioned.

polysiloxane and an aryl alcohol derivative is preferred from a point of heat resistance and an electrical propriety.

resin. A bisphenol resin is manufactured by the reaction of epichlorohydrin with bisphenol A. The bisphenol A is manufactured by the polymerization of phenol and acryloyl chloride. The bisphenol A is a white, crystalline solid which is soluble in organic solvents. It is used as a monomer in the manufacture of epoxy resins. Epoxy resins are manufactured by the reaction of epichlorohydrin with bisphenol A. The bisphenol A is a white, crystalline solid which is soluble in organic solvents. It is used as a monomer in the manufacture of epoxy resins.

reaction, such as acrylic acid, a trihydroxy phenylmethane, tetra hydroxy ethane, and extracts of phenol, is mentioned, and these one half or more are used. When using an epoxy resin together in the reaction-of-degeneration thing to which it comes to carry out the reaction of degeneration of the silane coupling agent expressed with a polyisobutylene compound and the above-mentioned general formula (1), the reaction-of-degeneration thereon. As opposed to the reaction-of-degeneration of the silane coupling agent (1) in the constituent of this invention, it is the reaction-of-degeneration thereon. As opposed to the reaction-of-degeneration of the silane coupling agent expressed with a polyisobutylene compound and the above-mentioned general formula (1). In the constituent of this invention, the total weight of an epoxy resin and a hardening agent — $25 \text{--} 300 \text{ weight percent}$, the total weight of an epoxy resin and a hardening agent — $25 \text{--} 300 \text{ weight percent}$ preferably, moreover, — the rate of an epoxy resin and a hardening agent mixed

The epoxy resin --- a hardening agent --- the range of 0.5-2.0 preferably.

melting nature is preferred. The mixture of the silica powder at the time of shaping or molding is a glassy form or a globular form, and an infinite time is preferred. If the thermoeetting resin 100 weight percent section which makes a reaction or degeneration takes a subject, it is required to be 100-400 weight percent section; and the hardness of an inorganic bulking agent are 200-600 preferably, it is a weight section. The above-mentioned inorganic bulking agent may deform the surface beforehand by finishing agents, such as the Slant system, a flame system, an aluminate system, and a zirconium system coupling agent. As far as from the slant coupling agent used for this invention and occasion demands, the Slant system

glycidoxypromethylmethoxysilane, 2-(3,4-epoxybutyl) methylmethoxysilane, 3-methacryloxypropylmethoxysilane, 3-mercaptopropylmethoxysilane, etc. can be mentioned, and these one kind or two kinds or more are used.

phosphine; tetraphenyl phosphonium tetrakisphenylborate, tetraphenylboron salts, such as triphenyl boron.
<http://www4.indiapti.go.id/cgi-bin/tranweb.cgi?clis?atw> <https://www4.jpdt.inpit.gov>... 2010/10/10.

tetrahexyl borate; the 1,8-diaza-bicyclo (5.4.0) undecene 7 and its derivative 7 and its derivative are mentioned. These hardening accelerators may be used independently, may use two or more kinds together, and can also use one or more kinds and/or a two compound together, if needed. The content of these hardening accelerators is used in the range of 0.01 to 10 weight section to thermosetting resin [0017].

[Working example] Hereafter, an embodiment explains this invention concretely. In an embodiment, the test method of the performance of a constituent is as follows.

- glass-transition-temperature: — TMA method and bending strength: — US K-6911, and solder immersion test: — immediately after heating 25 semiconductor devices for an examination to 85 °C and 85% constant temperature/humidity chamber for 168 hours, it is immersed in a 260 °C molten solder bath for 10 seconds.

Then, the number of the semiconductor devices in which the crack generated in package resin was counted.

The damp-proof test after solder immersion: In a solder immersion test, a crack of the semiconductor device cooker tester, electrical continuity is checked for every fixed time, and the poor incidence rate by the corrosion of aluminum wing measure time to reach to 50%.

[0018] Example A-G of manufacture (manufacture of reaction-of-degeneration thing A-G)

The polyimide compound was heated in the reaction vessel equipped with an agitator, a thermometer, and a condenser, it warmed at 160 °C, the silane coupling agent was heated so that it might become the presentation of A-G of the 1st table, and you made it reheat for 3 minutes, it cooled immediately, and reaction-of-degeneration thing A-G was obtained.

[0019] Example H-K of manufacture (manufacture of reaction-of-degeneration thing H-K)

A polyimide compound and phenol resin are inserted in the reaction vessel equipped with an agitator, a thermometer, and a condenser, and it warms at 160 °C, and is the 1st table. The silane coupling agent was inserted so that it might become the presentation of H-K of [Table 1], and you made it reheat for 5 minutes, it cooled immediately, and reaction-of-degeneration thing A-G was obtained.

[Table 1]

実験区別	ポリマー	ジシランカーブ (重合率) (重量率)	ノモルカーブ (重合率) (重量率)
A	#777-1 F 化合物(1)	KBM6 0 3 5	
B	#777-1 F 化合物(1)	KBM6 0 2 5	
C	#777-1 F 化合物(1)	SZ-6 0 8 3 5	
D	#777-1 F 化合物(1)	A-1 8 7 5	
E	#777-1 F 化合物(2)	KBM6 0 3 5	
F	#777-1 F 化合物(2)	SZ-6 0 8 3 5	
G	#777-1 F 化合物(2)	A-1 8 7 5	
H	#777-1 F 化合物(1)	KBM6 0 3 5	20.7
I	#777-1 F 化合物(1)	SZ-6 0 8 3 5	20.7
J	#777-1 F 化合物(2)	KBM6 0 3 5	20.7
K	#777-1 F 化合物(2)	SZ-6 0 8 3 5	20.7

[0021] Embodiment 1-G and the comparative example 1—the 8 2nd table The compound of the presentation (weight section) shown in [Table 2] was mixed with the Harashel river, and melting and the ground, were done for 3 minutes with a furher 100-130 °C hot calender roll. This mixture was cooled and melted, was tabletted and the molding resin composition was obtained. The following were used for the raw material used all over the 1st and 2nd table.

— Polyimide compound (1); bis-(4-maleimide phenyl)methane (product made from Mitsui Toatsu Chemicals (Chemistry))

— Polyimide compound (2); 2-(4-maleimide phenoxy)bis(3-maleimide phenyl) triphenoxyane (KBM603, Shin-Etsu Chemicals, Inc.)

— Epoxy resin; oxetane novolak type epoxy resin (OCN-1020, Nippon Kayaku Co., Ltd. make)

— Boron-fused epoxy resin; boron-fused phenol novolak type epoxy resin (BREN-S, Nippon Kayaku Co., Ltd. make)

— Hardening agent; novolac type phenol resin (PN-80, Nippon Kayaku Co., Ltd. make)

— Inorganic bulking agent (1); globular form used silica with a mean particle diameter of 20 micro (Hari Mick S-OO, Micron Make)

— Inorganic bulking agent (2); infinite form fused silica with a mean particle diameter of 13 micro (made in fuge REXX RD-8 (taumon))

— Silica coupling agent; 3-(2-aminoethyl aminoethyl) trimethoxysilane (KBM603, Shin-Etsu Chemicals Co., Ltd. make)

— Silane coupling agent; 3-(2-aminoethyl aminoethyl) propyl trimethoxysilane (KBM602, Shin-Etsu Chemicals Co., Ltd. make)

- Silane coupling agent: *p*-phenylenimine propyltrimethoxysilane (S2-1083; Dow Corning Toray Silicone make)
- Silane coupling agent: 3-allyldiisopropylmethoxysilane (A-187; Nippon Unicar make)
- Foresight auxiliary agent: antimony oxide; made by Sumitomo Metal Mining Co., Ltd.)
- Hardening accelerator (C₁₇Z₂; made in Shikoku Chemicals)

[Table 2] [0022]

	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	100	101	102	103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118	119	120	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135	136	137	138	139	140	141	142	143	144	145	146	147	148	149	150	151	152	153	154	155	156	157	158	159	160	161	162	163	164	165	166	167	168	169	170	171	172	173	174	175	176	177	178	179	180	181	182	183	184	185	186	187	188	189	190	191	192	193	194	195	196	197	198	199	200	201	202	203	204	205	206	207	208	209	210	211	212	213	214	215	216	217	218	219	220	221	222	223	224	225	226	227	228	229	230	231	232	233	234	235	236	237	238	239	240	241	242	243	244	245	246	247	248	249	250	251	252	253	254	255	256	257	258	259	260	261	262	263	264	265	266	267	268	269	270	271	272	273	274	275	276	277	278	279	280	281	282	283	284	285	286	287	288	289	290	291	292	293	294	295	296	297	298	299	300	301	302	303	304	305	306	307	308	309	310	311	312	313	314	315	316	317	318	319	320	321	322	323	324	325	326	327	328	329	330	331	332	333	334	335	336	337	338	339	340	341	342	343	344	345	346	347	348	349	350	351	352	353	354	355	356	357	358	359	360	361	362	363	364	365	366	367	368	369	370	371	372	373	374	375	376	377	378	379	380	381	382	383	384	385	386	387	388	389	390	391	392	393	394	395	396	397	398	399	400	401	402	403	404	405	406	407	408	409	410	411	412	413	414	415	416	417	418	419	420	421	422	423	424	425	426	427	428	429	430	431	432	433	434	435	436	437	438	439	440	441	442	443	444	445	446	447	448	449	450	451	452	453	454	455	456	457	458	459	460	461	462	463	464	465	466	467	468	469	470	471	472	473	474	475	476	477	478	479	480	481	482	483	484	485	486	487	488	489	490	491	492	493	494	495	496	497	498	499	500	501	502	503	504	505	506	507	508	509	510	511	512	513	514	515	516	517	518	519	520	521	522	523	524	525	526	527	528	529	530	531	532	533	534	535	536	537	538	539	540	541	542	543	544	545	546	547	548	549	550	551	552	553	554	555	556	557	558	559	560	561	562	563	564	565	566	567	568	569	570	571	572	573	574	575	576	577	578	579	580	581	582	583	584	585	586	587	588	589	590	591	592	593	594	595	596	597	598	599	600	601	602	603	604	605	606	607	608	609	610	611	612	613	614	615	616	617	618	619	620	621	622	623	624	625	626	627	628	629	630	631	632	633	634	635	636	637	638	639	640	641	642	643	644	645	646	647	648	649	650	651	652	653	654	655	656	657	658	659	660	661	662	663	664	665	666	667	668	669	670	671	672	673	674	675	676	677	678	679	680	681	682	683	684	685	686	687	688	689	690	691	692	693	694	695	696	697	698	699	700	701	702	703	704	705	706	707	708	709	710	711	712	713	714	715	716	717	718	719	720	721	722	723	724	725	726	727	728	729	730	731	732	733	734	735	736	737	738	739	740	741	742	743	744	745	746	747	748	749	750	751	752	753	754	755	756	757	758	759	760	761	762	763	764	765	766	767	768	769	770	771	772	773	774	775	776	777	778	779	780	781	782	783	784	785	786	787	788	789	790	791	792	793	794	795	796	797	798	799	800	801	802	803	804	805	806	807	808	809	810	811	812	813	814	815	816	817	818	819	820	821	822	823	824	825	826	827	828	829	830	831	832	833	834	835	836	837	838	839	840	841	842	843	844	845	846	847	848	849	850	851	852	853	854	855	856	857	858	859	860	861	862	863	864	865	866	867	868	869	870	871	872	873	874	875	876	877	878	879	880	881	882	883	884	885	886	887	888	889	890	891	892	893	894	895	896	897	898	899	900	901	902	903	904	905	906	907	908	909	910	911	912	913	914	915	916	917	918	919	920	921	922	923	924	925	926	927	928	929	930	931	932	933	934	935	936	937	938	939	940	941	942	943	944	945	946	947	948	949	950	951	952	953	954	955	956	957	958	959	960	961	962	963	964	965	966	967	968	969	970	971	972	973	974	975	976	977	978	979	980	981	982	983	984	985	986	987	988	989	990	991	992	993	994	995	996	997	998	999	1000	1001	1002	1003	1004	1005	1006	1007	1008	1009	1010	1011	1012	1013	1014	1015	1016	1017	1018	1019	1020	1021	1022	1023	1024	1025	1026	1027	1028	1029	1030	1031	1032	1033	1034	1035	1036	1037	1038	1039	1040	1041	1042	1043	1044	1045	1046	1047	1048	1049	1050	1051	1052	1053	1054	1055	1056	1057	1058	1059	1060	1061	1062	1063	1064	1065	1066	1067	1068	1069	1070	1071	1072	1073	1074	1075	1076	1077	1078	1079	1080	1081	1082	1083	1084	1085	1086	1087	1088	1089	1090	1091	1092	1093	1094	1095	1096	1097	1098	1099	1100	1101	1102	1103	1104	1105	1106	1107	1108	1109	1110	1111	1112	1113	1114	1115	1116	1117	1118	1119	1120	1121	1122	1123	1124	1125	1126	1127	1128	1129	1130	1131	1132	1133	1134	1135	1136	1137	1138	1139	1140	1141	1142	1143	1144	1145	1146	1147	1148	1149	1150	1151	1152	1153	1154	1155	1156	1157	1158	1159	1160	1161	1162	1163	1164	1165	1166	1167	1168	1169	1170	1171	1172	1173	1174	1175	1176	1177	1178	1179	1180	1181	1182	1183	1184	1185	1186	1187	1188	1189	1190	1191	1192	1193	1194	1195	1196	1197	1198	1199	1200	1201	1202	1203	1204	1205	1206	1207	1208	1209	1210	1211	1212	1213	1214	1215	1216	1217	1218	1219	1220	1221	1222	1223	1224	1225	1226	1227	1228	1229	1230	1231	1232	1233	1234	1235	1236	1237	1238	1239	1240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